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Waveform-sequencing for scanning tunneling microscopy based pump-probe spectroscopy and pulsed-ESR

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Abstract: We present here the straightforward implementation of pump-probe methods into existing scanning tunneling microscopy (STM) systems. Our method uses the waveform-sequencing abilities of a standard arbitrary waveform generator (AWG) and a simple mechanical relay switch that either connects the regular STM control electronics or the AWG to the STM system. Our upgrade further enables pulsed-ESR excitation for advanced STM based spin-resonance experiments. We demonstrate the technical implementation, signal detection using a lock-in amplifier, and cross-correlation measurements of DC/DC and DC/RF pulses highlighting our ~ 5 ns time resolution, here limited by the speed of the available electronics. Our setup is highly versatile and can be extended to suit other needs of STM based investigations such as required in diverse mapping schemes or the coherent manipulation of qubits. • Plug and Play Pump-Probe Capability • One setup for Pump-Probe Spectroscopy and Pulsed-ESR • Waveform Memory Saving and Versatile due to Waveform-Sequencing of Arbitrary Waveforms

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Method Article

Waveform-sequencing for scanning tunneling microscopy based pump-probe spectroscopy and pulsed-ESR



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A B S T R A C T

We present here the straightforward implementation of pump-probe methods into existing scanning tunneling microscopy (STM) systems. Our method uses the waveform-sequencing abilities of a standard arbitrary waveform generator (AWG) and a simple mechanical relay switch that either connects the regular STM control electronics or the AWG to the STM system. Our upgrade further enables pulsed-ESR excitation for advanced STM based spin-resonance experiments. We demonstrate the technical implementation, signal detection using a lock-in amplifier, and cross-correlation measurements of DC/DC and DC/RF pulses highlighting our 5 ns time resolution, here limited by the speed of the available electronics. Our setup is highly versatile and can be extended to suit other needs of STM based investigations such as required in diverse mapping schemes or the coherent manipulation of qubits.

- Plug and Play Pump-Probe Capability
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A R T I C L E I N F O

Method name: Waveform-sequencing for scanning tunneling microscope based pump-probe spectroscopy and pulsed-ESR

Keywords: Scanning tunneling microscopy, Pump-probe spectroscopy, Pulsed-ESR, Time resolved measurements

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More specific subject area:	Surface physics, time-resolved measurements, relaxation dynamics
Method name:	Waveform-sequencing for scanning tunneling microscope based pump-probe spectroscopy and pulsed-ESR
Name and reference of original method:	Measurement of the Fast Electron Spin Relaxation Times with Atomic Resolution, Loth et al., Science 329 , 1628 (2010)
Resource availability:	NA

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Method details

Broader context

The small signals and the concomitantly required amplification through transimpedance amplifiers limit the bandwidth of scanning tunneling microscopes (STM) to the kilohertz range. Although specialized circuit designs may run at MHz frequencies [1], they operate at cryogenic temperatures which renders them difficult to implement and they achieve only small bandwidths, thus limiting their versatility.

The introduction of pump-probe techniques for STM systems by Nunes and Freeman [2,3] more than two decades ago lifted amplifier related constraints of the bandwidth and promised unprecedented access to the temporal dynamics of atomic scale systems. However, these earlier approaches and more recent ones using THz pulses [4,5] require either dedicated sample and tip designs or a sophisticated external setup for creating pulses and their coupling into the STM junction.

The elegant all-electronic pump-probe spectroscopy method, introduced by Loth et al. [6], enabled investigation of spin relaxation times in the nanoseconds range with atomic resolution. It has since served to measure the dynamics of long-lived antiferromagnetic structures [7,8] and individual atoms [6,9–13]. In these pump-probe setups [6,14,15] the existing bias voltage line of an STM, dedicated pulse pattern generators, external power combiners, and an RF relay acting as a chopper to modulate the probe-pulses for lock-in detection are used. It is surprising that even though these solutions appear quite straightforward, they have not seen a more widespread implementation by the community.

We therefore introduce here an even simpler setup for obtaining pump-probe capabilities in the nanoseconds range using only a standard arbitrary waveform generator (Keysight 33600A) and its waveform-sequencing function. Our approach is truly *plug-and-play*, affordable, and should alleviate any remaining hesitations encountered by other researchers in the planning of such an addition to their existing STM apparatus.

Method details

The integration of pump-probe capabilities only briefly interrupts the existing setup in which we add the in-line transfer relay to the bias voltage path [16]. The schematics in Fig. 1 shows the hookup of the AWG (red) to the STM control electronics and the optional RF generator (dashed yellow). In contrast to previous pump-probe setups where an RF relay served as a chopper for lock-in detection, we here use a mechanical relay as a transfer switch to either connect the DC bias from the STM control electronics (for tunneling spectroscopy and regular topographic imaging) or the pulses from the AWG to the STM apparatus [16]. For the present purpose, we use a PCB mount relay (Tianbo) that we control with a 5 V USB power supply and to which we solder BNC connectors. Different relays of any sophistication may equally work but are not needed in our case, since neither edge times are affected nor is any ringing introduced by the relay.

Our method for pump-probe spectroscopy and pulsed-ESR uses the waveform-sequencing ability of the AWG. Waveform-sequencing is essentially the consecutive and seamless execution of a list of predefined arbitrary waveforms (arbs), often compared to playing songs from a playlist in a music player.

Fig. 2a illustrates our method. We first create three arbs for the pump-pulse (arbP: yellow) and probe-pulses (arbA: green, and arbB: orange) using MATLAB. These arbs have equal sample length of $l = t_{arb} \times SR$, where SR is the sample-rate of the AWG. The length of the arbs should be designed with the dynamics of the studied system in mind. This means that the total duration of the arb, i.e., t_{arbA} , should be several times the expected relaxation times to ensure excitation of the system from a known equilibrium state. The two distinct probe-pulse arbs (arbA and arbB) are applied cyclically in Ch1 at about $f_{cycle} = 887$ Hz to modulate the tunneling current for lock-in detection. Our waveform playlist is composed of a dedicated arb sequence of probe-pulses consisting of N segments of arbA (cycle A) followed by N segments of arbB (cycle B), where $N = \left\lceil 1/2f_{cycle}t_{arbA} \right\rceil$. To provide a synchronization

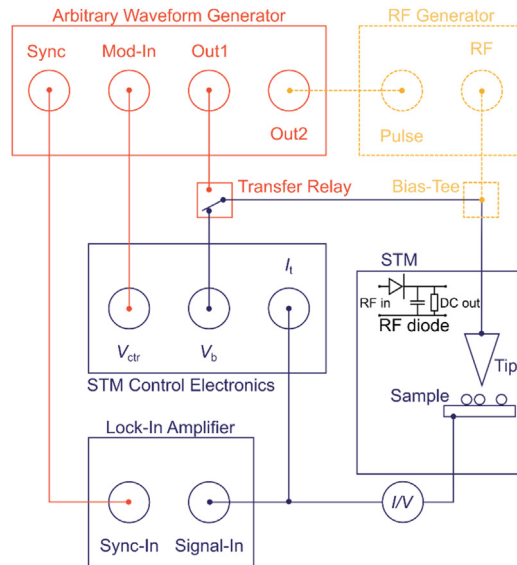


Fig. 1. Blue drawings indicate existing STM system, control electronics, and lock-in amplifier. The red sketch shows how to implement the AWG into that system using a transfer relay. The dashed yellow drawing is the optional integration of a pulsed RF generator. The inset in the STM box indicates the RF diode circuit used to emulate an STM in the present work.

signal for the lock-in amplifier, we set the sync output to high (Hi) during cycle A, and during cycle B we set it to low (Lo). The effective f_{cycle} will depend on the rounding down in the determination of N .

The execution of the pump-arb is simpler as we play it perpetually on Ch2, as also indicated in Fig. 2a. The waveform-sequence of Ch1 and the pump-arbs on Ch2 are added internally by the AWG and finally applied to output 1. As an example, we show in Fig. 2b a measured time-trace of this combined output 1 signal, recorded with an oscilloscope at the threshold between cycle A and B. The transition from one cycle to the other is as seamless as the transitions between any two arbs. The three arbs used here have a duration of 50 ns. ArbP contains a 5 ns wide pulse of 150 mV amplitude and is generated by Ch2. The waveform-sequence composed of arbA and arbB is generated on Ch1 and contains a 10 ns wide pulse of 50 mV amplitude for arbA, while arbB = −arbA.

We now discuss how to use this setup for time-resolved measurements. As we see from Fig. 2b, the pulses constitute only a short part of their respective arb. Since our waveform-sequencing guarantees the perfect synchronization of Ch1 and Ch2, arbP will always be in sync with arbA and arbB. This leaves the pulse-position within the arb, i.e., the phase, as the handle through which to control the time-delay Δt between pump and probe-pulses.

We conveniently vary the position of the pump-pulse within arbP via the phase modulation function (PM, see Fig. 3a). To that end, we use the Mod-In input of the AWG to steer the phase of Ch2 by using an external DC voltage (V_{ctr}) from the STM control electronics (see schematics Fig. 1). A dedicated DAQ card may serve that purpose as well. We adjust the phase of arbP by ± 180 degrees for $V_{\text{ctr}} = \pm 5$ V. This is equivalent to the circular shift of the pulse position within arbP when V_{ctr} is swept from -5 to $+5$ V and corresponds to a variation of the relative time Δt between pump and probe-pulse in the fully available range t_{arbA} . Note that the phase of Ch2 may also be adjusted via the computer control of the AWG, which might be useful for certain applications or when no control voltage is available.

Similarly, for a chosen time delay between pump and probe-pulse, we again use the amplitude modulation (AM, see schematics in Fig. 3b) of the AWG to control the amplitude of the pump-pulses via V_{ctr} . A voltage of $+5$ V corresponds to the maximal amplitude and -5 V to the maximal negative amplitude. The AM mode is useful in the search for threshold voltages at which dynamical processes are excited, as for instance shown in previous work [6,9].

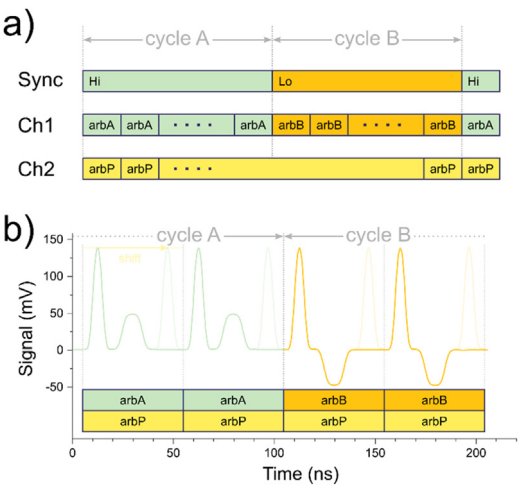


Fig. 2. Waveform-sequencing concept. a) The waveform-sequence consist of two repeated cycles of distinct probe-arbs (arbA and arbB) and is played on Ch1. The pump-arb (arbP) is generated in Ch2. Both channels are added internally by the AWG and applied to output 1. A sync-signal marks the temporal position within the waveform-sequence for lock-in detection. b) Measured time-trace of output 1 at the end of cycle A, demonstrating the seamless transition between the cycles. The shift of the probe-pulse is used in time-resolved measurements and serves to vary the time-delay between pump and probe-pulse.

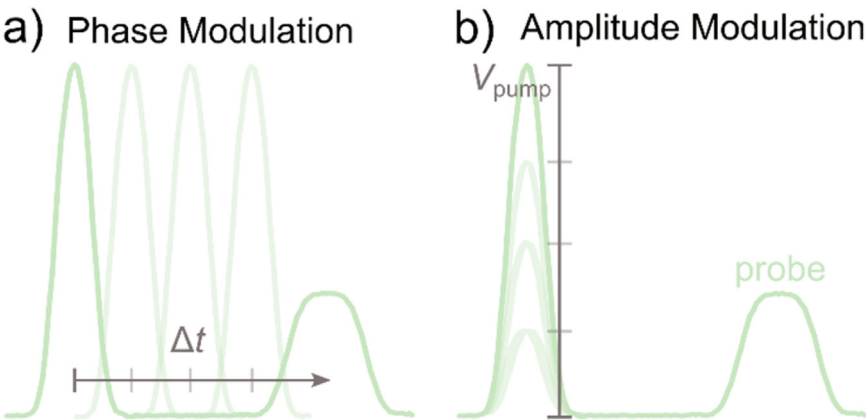


Fig. 3. Modulation schemes acting on the pump-arb (arbP), played on Ch2. (a) The phase modulation (PM) adjusts the relative time Δt between the pump and the probe-pulse. (b) The amplitude modulation (AM) controls the pump-pulse amplitude at fixed Δt . Both modes are controlled via the external V_{ctr} voltage.

Method validation

To reveal the time-resolution of our method through the bottle neck of a bandwidth limited system, for instance a transimpedance amplifier, we exploit a nonlinear tunneling junction, which is frequently used as a base for time-resolved measurements [2]. We simulate here a nonlinear tunneling junction by a Schottky diode (Pasternack, PE8014), indicated by the diode circuit in the inset of Fig. 1. The nonlinearity of the diode creates an additional current when pump and probe-pulses coincide due to $I(V_{pump} + V_{probe}) \neq I(V_{pump}) + I(V_{probe})$ [2]. Note that an actual STM might show signs of impedance

mismatches (reflections) that may have to be compensated, for instance via pulse-shaping described by Grosse and coworkers [17] or suppressed by adjusting the edge times. Fig. 4 shows the diode signal when we overlap pump and probe-pulses generated by the earlier described waveform-sequencing method and the phase-modulation. At first, we show the overlap of DC pump and DC probe-pulses on the nonlinearity in panel a. We vary the duration of the pump-pulse t_{pump} from 5 to 15 ns at a fixed probe-pulse width of 10 ns to reveal the correlation effect of the nonlinearity.

We further use the same method to superimpose RF pump-pulses and DC probe-pulses, as seen in the resulting correlation trace in Fig. 4b. Instead of internally combining Ch1 and Ch2 on output 1, we now feed Ch2 directly to the pulse-trigger input of the RF generator (Keysight N5183B). As indicated in Fig. 1, the cabling remains conveniently untouched for this operation mode. The output of Ch1 is now solely playing the waveform-sequence of probe-pulses (arbA, arbB). The launched RF pulses travel via the resistive bias-tee (Tektronix, PSPL5370) to the tunneling junction where they overlap with the DC pulses. For this demonstration, we set the generator to a frequency of 20 GHz at a power level of -5 dBm. The probe-pulse width t_{probe} is held constant in Fig. 4b while we vary the width of the triggered RF pulses t_{pump} from 5 ns to 25 ns. As can be seen in the correlation data, the width of the RF pulses scales with the width of their trigger pulses, thus presenting all requirements for pulsed-ESR methods that require the controlled application of RF pulses of precise timing and duration.

The shape of the correlation curves in Fig. 4 reveals important information about the effective time-resolution and offers valuable diagnostical insight into the system. For instance, the roundness of the correlation data is determined by the edge-times of the pump and probe-pulses, which is here about 3 ns and limited by the speed of our AWG. Further, the correlation data is useful in the detection of

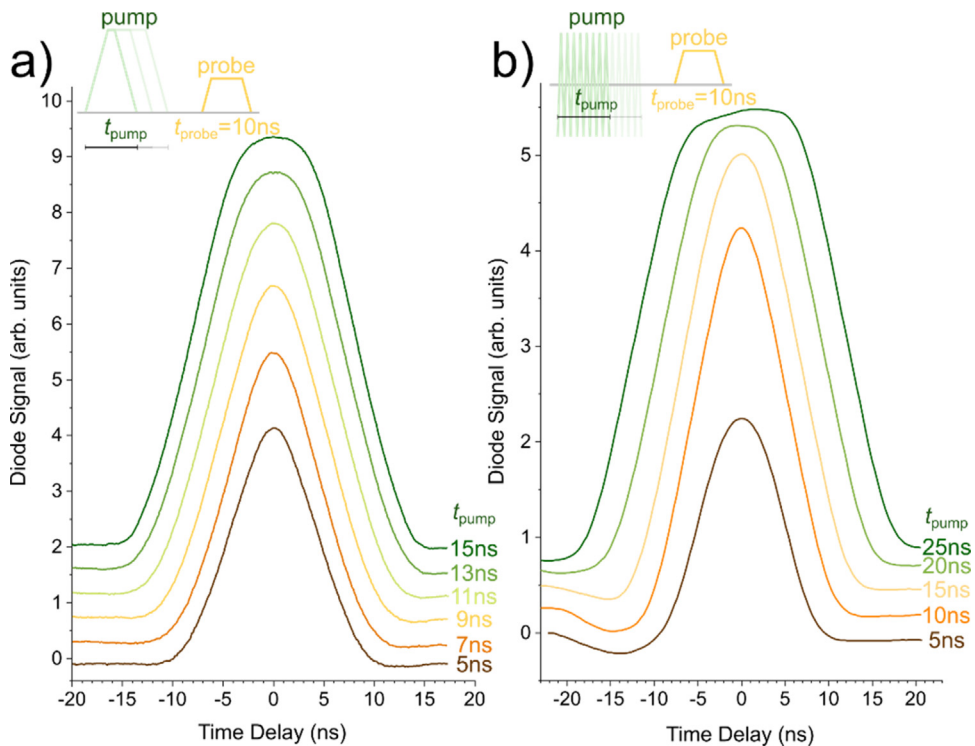


Fig. 4. Cross correlation of pulses measured via a nonlinearity using (a) DC pump with DC probe-pulses (10 ns) and (b) RF pump with DC probe-pulses (10 ns). The pump-pulse width is indicated next to the respective correlation traces. The cross-correlation trace in panel (b) shows a dip at negative time delays due to slight ringing introduced by the bias-tee, which has an impedance mismatch.

ringing due to the presence of impedance mismatches [17], which lead to the subtle dip in Fig. 4b and was caused by our mismatched bias-tee (pick-off impedance of $450\ \Omega$).

The time-delay of the maxima in the correlation data, such as in Fig. 4, is extremely sensitive to variations in the cable lengths. To estimate the minimal length to which we are sensitive, we add cable segments of different lengths to the Ch2-Pulse path (Fig. 1). The length-dependent time-delay of the correlation maxima is measured to a precision of 30 ps. From that data, we find the cable propagation time of $(4.98 \pm 0.06)\ \text{ns m}^{-1}$, a corresponding velocity factor of (0.671 ± 0.008) , and a sensitivity to variations in the cable length of about 6 mm.

Discussion and tweaks

The lock-in amplifier measures the difference signal $I_A - I_B$, that is, the difference between the signal created during cycle A and cycle B. This leads to the cancellation of the currents due to arbP, since the pump-pulse is the same in both parts of the cycle. Consequently, the interesting signal carrying the dynamics of the system due to the probe-pulse is enhanced. The earliest implementation of the all-electronic pump-probe method used a chopper, effectively suppressing arbB [6]. In our implementation, this would be equivalent to setting $\text{arbB} = 0$ during cycle B [6]. A better signal is achieved, through the inverted voltage of $\text{arbB} = -\text{arbA}$, because we obtain data also for the second half of the entire cycle. However, since the preamplifier acts like a charge storage that is loaded and unloaded through the voltage induced current pulses, it may become overloaded through the voltage pulses of arbP. A remedy is the application of current-cancelling pump-pulses, either via an AC pulse (as shown above using the RF generator) or a positive/negative pulse [18]. Our waveform-sequencing method can be easily modified to overcome these issues that may arise if the ratio of the pump and probe-pulse amplitudes becomes too big, by simply changing the form of arbP.

The waveform-sequencing methods described here allows for a flexible design of arbitrarily shaped pump/probe-pulse trains. Our method will also work, if pulse-shaping is employed to compensate for a pulse deformation due to frequency dependent transmission [17], since our method is completely independent of the explicit shape of the arb.

Waveform-sequencing may also help overcome memory related limitations. For instance, a waveform containing the entire cycle A and B inside a single waveform as done previously can occupy a large portion of the waveform memory and it needs to be transferred to the AWG during the experiment. This may not only lead to an interruption of the AWG output but the permanent data transfer for different time-increments also creates noticeable overhead. Depending on the AWG memory, explicitly coding of an entire cycle A and B, may further require prolonging effective edge-times. No such limitations are present in our method. We achieve rising and falling edge times of 3 ns for any pulse-width at full sample-rate of 1 GSa/s. The shortest pulse-width is limited here only by the technical characteristics of our AWG.

We should also mention that any two voltage sources will have different voltage offsets that need to be compensated. This is particularly important, because as we switch between two nominally equivalent DC voltages of the STM control electronics and the AWG, we might unknowingly apply a gate voltage during the relaxation phase of an experiment. We therefore calibrate the voltage offsets of AWG and STM electronics by separately measuring two current-voltage traces at two impedances (for instance $1\ \text{G}\Omega$ and $100\ \text{M}\Omega$). The crossing of the two I-V traces determines the zero-bias offset of the respective voltage source. As a verification, a correction of the offsets should lead to the same current or tip-height when switching the transfer relay from the STM control to the AWG. Note that we typically switch the relay in open feedback condition and while the tip is pulled back by 1 nm. The lifting of the tip was introduced as a safety measure after the observation of voltage spikes for some relay designs that may cause local tip or sample damage.

We foresee the utility of our setup also for the anticipated advent of STM based pulsed-ESR methods. This is owed to the tremendous promise revealed in the study of individual electron and nuclear spins [12, 19–21] through the introduction of ESR capable STM systems [16,22]. Although these STM based ESR studies relied on continuous wave excitation, the ongoing search for spin systems having long coherence times or the exploitation of clock-transitions [23–25] will ultimately lead to the introduction of pulsed ESR-STM methods that coherently control the quantum states of atomic and molecular qubits.

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